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Submitted for publication in

Synthetic Metals

Proceedings of the International Conference on Synthetic Metals

Shanghai, China

June 29-July 5, 2002

The submitted manuscript has been created by the University of Chicago as Operator of Argonne National Laboratory ("Argonne") under Contract No. W-31-109-ENG-38 with the U.S. Department of Energy. The U.S. Government retains for itself, and others acting on its behalf, a paid-up, nonexclusive, irrevocable worldwide license in said article to reproduce, prepare derivative works, distribute copies to the public, and perform publicly and display publicly, by or on behalf of the Government.

*Work supported by the U.S. Department of Energy, BES-Materials Sciences, under Contract W-31-109-ENG-38.

Unusual interlayer transport in quasi-two-dimensional organic metals

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Abstract

The interlayer transport properties of the organic superconductor $\beta''\text{-(ET)}_2\text{SF}_5\text{CH}_2\text{CF}_2\text{SO}_3$ are presented. The resistivity perpendicular to the highly conducting ET layers is about 100 times larger than compared to other quasi-two-dimensional ET salts. For a magnetic field parallel to the layers no peak could be resolved in the angle-dependent interlayer resistance which proved the coherent nature of transport in other ET salts. This and the absence of any further clear-cut proof for a coherent quasiparticle motion suggest an incoherent nature of the interlayer transport in $\beta''\text{-(ET)}_2\text{SF}_5\text{CH}_2\text{CF}_2\text{SO}_3$.

Keywords: Organic superconductors, Transport measurements

An understanding of the normal-state properties is an essential task in order to better understand also the superconductivity in organic metals. In the quasi-two-dimensional (2D) organic conductors based on the donor ET (= bisethylenedithio-tetrathiafulvalene) the electronic properties seem to follow a well-behaved Fermi-liquid description. This appears to be evident by the often observed magnetic quantum oscillations characteristic for a well-developed Fermi surface. However, if there are no beating nodes in the oscillating signal present, the interlayer band structure leaves unclear. In the case when the interlayer transport is incoherent, i.e., when an electron loses his phase information between successive tunneling processes, no Bloch states can evolve and the band picture fails, i.e., a Fermi surface is only defined within the layers [1].

Incoherent interlayer transport is expected when the intralayer scattering rate η/τ is much larger than the interlayer transfer integral t_c . As we have shown recently, this so-called Mott-Ioffe-Regel would predict incoherent transport for the 2D organic metal $\kappa\text{-(ET)}_2\text{I}_3$ when τ is estimated from the experimentally determined Dingle temperature of about 0.25 K [2]. Nevertheless, a clear-cut proof for the coherent nature of the interlayer transport was evidenced by the observation of a peak in the angular-dependent interlayer resistance that appeared for magnetic fields aligned exactly along the ET planes. A similar observation was recently reported for the isostructural

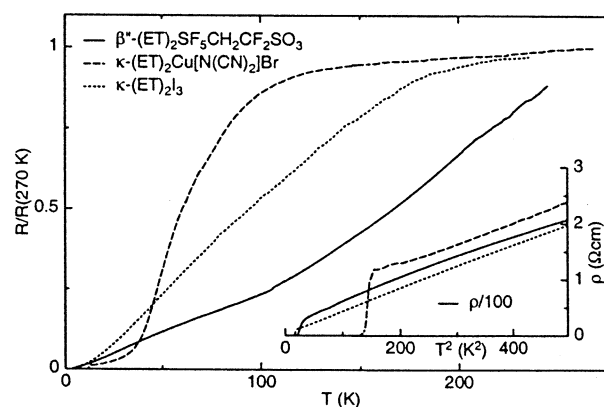


Fig. 1. Temperature dependence of the interlayer resistance R normalized by R at 270 K for three different organic superconductors. The inset shows the resistivity ρ as a function of T^2 . Note that ρ for $\beta''\text{-(ET)}_2\text{SF}_5\text{CH}_2\text{CF}_2\text{SO}_3$ has been divided by 100.

superconductor $\kappa\text{-(ET)}_2\text{Cu}(\text{NCS})_2$ [3]. These results indicate that the Mott-Ioffe-Regel based on τ obtained from a Dingle analysis should only be used as an order-of-magnitude estimate for possible incoherent transport.

Indeed, a qualitatively different behavior of the interlayer-transport properties occurs for the organic metal $\beta''\text{-(ET)}_2\text{SF}_5\text{CH}_2\text{CF}_2\text{SO}_3$. This becomes evident already from the absolute value of the resistivity ρ compared to other organic metals (Fig. 1). Although all the organic conductors

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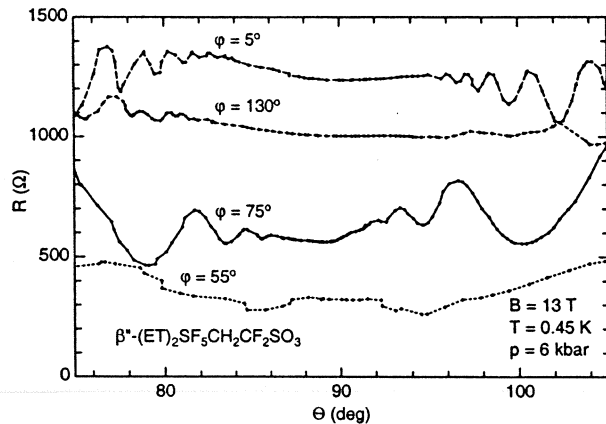


Fig. 2. Interlayer resistance close to 90 deg of a β'' -(ET) $_2\text{SF}_5\text{CH}_2\text{CF}_2\text{SO}_3$ sample under hydrostatic pressure of 6 kbar for different azimuthal angles φ .

show a metallic temperature dependence of the resistivity, starting from room temperature, no generic T dependence for the different materials exists. Nevertheless, towards lower T , ρ can fairly well be described by $\rho = \rho_0 + AT^2$ (inset of Fig. 1). Usually, this T^2 behavior is taken as a signature for a Fermi liquid where scattering is dominated by electron-electron interactions. However, the wide temperature range over which this T^2 dependence can be found and the extraordinary large value of the coefficient A is not explicable by the usual Landau theory for a Fermi liquid. A is by orders of magnitude larger than observed for any other metal including the strongly correlated heavy-fermion metals. It seems therefore highly questionable to seek the origin of the T^2 dependence solely in the electronic system. Scattering of the 2D quasiparticles at 3D phonons might be responsible for the observed feature.

Another important point to mention here, is the very high interlayer resistivity ρ of the organic metals. At low T , $\rho \approx 1 \Omega\text{cm}$ of κ -(ET) $_2\text{Cu}[\text{N}(\text{CN})_2]\text{Br}$ is about 10^6 times larger than ρ for good metals at room temperature (inset of Fig. 1). For β'' -(ET) $_2\text{SF}_5\text{CH}_2\text{CF}_2\text{SO}_3$, ρ is even larger by another factor of about 100. Within the usually applicable Bloch-Boltzmann transport theory the mean-free paths would be much smaller than interatomic distances, even at low T . Consequently one has to ask – especially pronounced for β'' -(ET) $_2\text{SF}_5\text{CH}_2\text{CF}_2\text{SO}_3$ – whether the usual Fermi-liquid theory adequately describes the interlayer transport.

According to Ref. [1], there exists no unequivocal proof for the proposed incoherent transport mechanism. There are, however, three unambiguous checks to test for coherent interlayer transport. All three tests failed for β'' -(ET) $_2\text{SF}_5\text{CH}_2\text{CF}_2\text{SO}_3$. First, no beats in the magnetic quantum oscillations are observable for fields starting at 60 T down to about 1.4 T, the lowest field to which these oscillations could be resolved. Second, no crossover from a linear to a quadratic field dependence of the interlayer resistance in a parallel magnetic field could be observed up to 28 T [2]. The resistance for that field orientation is growing even less than linear with field. However, one should note that much higher fields might be necessary for a valid test, since even

in metals with proven coherent transport this crossover has not been observed yet. The third signature proving coherent transport is the occurrence of a peak in the angular-dependent resistance at $\Theta = 90^\circ$, i.e., for fields applied parallel to the ET layers. Experimental results in search for this feature are shown in Fig. 2. No indication of a peak at 90° is found.

In Fig. 2 we show results for a sample investigated under hydrostatic pressure of about 6 kbar. This pressure was determined by monitoring T_c of a Sn wire mounted next to the sample inside the CuBe pressure cell. For this pressure, T_c of β'' -(ET) $_2\text{SF}_5\text{CH}_2\text{CF}_2\text{SO}_3$ is reduced to below 0.5 K and, consequently, the field of 13 T, supplied by a superconducting magnet, is sufficient to completely reach the normal state even for $\Theta = 90^\circ$ and $T = 0.45$ K. In Fig. 2 only four out of many different azimuthal angles φ are shown. For all φ , all magnetic fields, all pressures, and all samples investigated never a peak at $\Theta = 90^\circ$ occurred. From this wealth of data we estimate that a possible peak must be smaller than about 0.001° . (The resistance was continuously monitored when the samples were rotated.) By using the equation $\Theta_{\text{peak}/2} = t_c c' k_F / \epsilon_F$ derived in [4], with $\Theta_{\text{peak}/2}$ the half width of the peak, c' the interlayer distance, k_F the Fermi wave vector, and ϵ_F the Fermi energy, the interlayer transfer integral must be smaller than about 10^{-7} eV. This is by about three orders of magnitude smaller than $t_c \approx 60 \mu\text{eV}$ obtained for the 2D metal κ -(ET) $_2\text{I}_3$ [2].

All these experimental tests, therefore, gave *no* hint whatsoever for a Bloch-Boltzmann-like coherent interlayer transport. Accordingly, β'' -(ET) $_2\text{SF}_5\text{CH}_2\text{CF}_2\text{SO}_3$ can be regarded as a perfect 2D metal. This is corroborated by the observation of inverse-sawtooth-like de Haas-van Alphen oscillations which perfectly fit the theoretical prediction for a 2D metal with fixed chemical potential [5]. Further on, for fields close to $\Theta = 0^\circ$ a field-induced metal-insulator transition and a violation of Kohler's rule is found [6]. This additional deviation from the conventional Bloch-Boltzmann transport theory strongly suggests that the electronic transport perpendicular to the layers most probably is incoherent in β'' -(ET) $_2\text{SF}_5\text{CH}_2\text{CF}_2\text{SO}_3$. Nevertheless, within the planes the Fermi-liquid picture is adequate since a well-defined Fermi surface can be resolved.

References

- [1] R.H. McKenzie, P. Moses, Phys. Rev. Lett. 81 (1998) 4492.
- [2] J. Wosnitzer, J. Hagel, J.S. Qualls, J.S. Brooks, E. Balthes, D. Schweitzer, J.A. Schlueter, U. Geiser, J. Mohtasham, R.W. Winter, G.L. Gard, Phys. Rev. B 64 (2002) 180506(R).
- [3] J. Singleton, P.A. Goddard, A. Ardavan, N. Harrison, S.J. Blundell, J.A. Schlueter, A.M. Kini, Phys. Rev. Lett. 88 (2002) 037001.
- [4] N. Hanasaki, S. Kagoshima, T. Hasegawa, T. Osada, N. Miura, Phys. Rev. B 57 (1998) 1336.
- [5] J. Wosnitzer, S. Wanka, J. Hagel, E. Balthes, N. Harrison, J.A. Schlueter, A.M. Kini, U. Geiser, J. Mohtasham, R.W. Winter, G.L. Gard, Phys. Rev. B 61 (2000) 7383.
- [6] J. Wosnitzer, S. Wanka, J. Hagel, H. v. Löhneysen, J.S. Qualls, J.S. Brooks, E. Balthes, J.A. Schlueter, U. Geiser, J. Mohtasham, R.W. Winter, G.L. Gard, Phys. Rev. Lett. 86 (2001) 508.